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ELECTROCHEMICAL AND GRAVIMETRIC INVESTIGATION OF THE CORROSION INHIBITION PERFORMANCE OF CLINDAMYCIN FOR MILD STEEL IN ACIDIC MEDIA

The corrosion inhibition performance of clindamycin for mild steel in 2.0 M H_2SO_4 has been studied using gravimetric and electrochemical methods. The results showed that the clindamycin inhibited mild steel corrosion in 2.0 M H_2SO_4 solution. The inhibition efficiency was found to increase with an increase in the concentration of clindamycin and decrease with an increase in the temperature. The maximum inhibition efficiency was obtained at 303 K (91.1%). Potentiodynamic polarization results reveal that clindamycin is a cathodic-type corrosion inhibitor lower concentrations and an anodic-type corrosion inhibitor at higher concentrations. The experimental data fitted the Langmuir adsorption isotherm and the adsorption of clindamycin was found to be spontaneous. The values of activation energy and Gibb's free energy were found within the range of limits expected for the mechanism of physical adsorption.

Keywords: Mild steel; corrosion; inhibition; sulphuric acid; clindamycin

1. Introduction

Corrosion is a naturally occurring phenomenon commonly defined as the deterioration of material by interaction with its environment. Corrosion of metals has caused huge economic losses involving billions of dollars each year in many industries. The international measure of prevention, application, and economics of corrosion technology (IMPACT) estimated the global cost of corrosion to be $2.5 trillion which is equivalent to 3.4% of gross domestic product GDP [1-2]. The IMPACT found that the introduction of corrosion prevention strategies could result in global savings between 15-35% of the cost damage. Therefore, control measures need to be implemented to reduce or inhibit corrosion thereby prolonging the life span of metals [2-5].

Several approaches have been suggested and implemented to protect metal against corrosion, one of which is the use of corrosion inhibitors. Although, the use of corrosion inhibitor is one of the best methods of controlling corrosion [5-9], most corrosion inhibitors are toxic, expensive, and not readily available. Thus, researchers have focused on the use of eco-friendly compounds that could be obtained conveniently from natural biomass [8-11]. These compounds normally contain electronegative atoms such as nitrogen, sulfur, and oxygen in their molecular structure. Presently a few nontoxic organic compounds including various drugs such as Amoxicillin [12], nifedipine [13], Ciprofloxacin [14], Amoldipine [15], Gentamicine and sulfamethoxazole [16], Irbesartan [17], Cephapirin [18], among others [19,20] have been reported to effectively retard corrosion rates. However, there is still a need for an extended research on other organic compounds that can offer comparable performance to the currently used toxic inhibitors for industrial applications.

Clindamycin is an antibiotic drug under the class of chloroquinolones. The IUPAC nomenclature of the drug is (2S,4R))−N−{(1S,2S)−2−chloro─1−{(2R,3R,4R,5R,6R)-3,4,5−trihydroxy−6−methylsulfanyloxan−2−yl}pr propyl}−1−methyl−4−propyrrolidine −2 – carboxamide. It has a molecular formula $C_{18}H_{33}ClN_2O_5S$ and a molecular weight of 424.98 g/mol. Clindamycin is rich in oxygen, sulfur, and nitrogen in addition to its heterocyclic rings which are appealing features expected of a potential corrosion inhibitor. The structure of clindamycin is shown in Fig. 1 [21].

Given that industrial acid-pickling processes are mostly carried out using acids, inhibitors are indispensable during these processes to reduce corrosion damage. Herein we present

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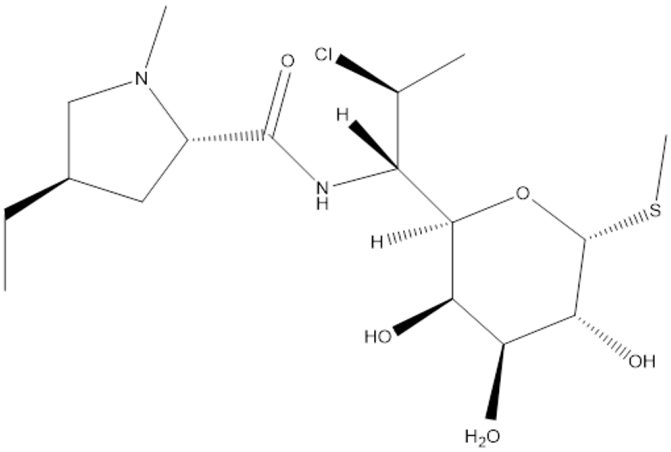

Fig. 1. Chemical structure of clindamycin drug

a cost-effective, ecofriendly, benign, readily available corrosion inhibitor (clindamycin) for mild steel in the presence of aggressive acidic environment. This present work aims to evaluate the corrosion inhibitive performance of clindamycin for mild steel in 2.0 M H_2SO_4 using the gravimetric and electrochemical techniques.

2. Materials and method

2.1. Clindamycin

Clindamycin powder was obtained from clindamycin capsules procured directly from Fidson pharmaceutical company, Lagos, Nigeria. Different concentrations of the clindamycin were prepared by dissolving appropriate quantities of the powder in the blank acid solution [22].

2.2. Corrosion medium

The stock solution (1000 ppm) of the inhibitor was prepared by dissolving 1 gram of clindamycin powder in 1.0 dm^3 of an already prepared 2.0 M H_2SO_4 solution (blank solution). Other desired concentrations ranging from 100 ppm to 500 ppm were prepared by serial dilution of the stock solution with distilled water.

2.3. Mild steel specimen

Mild steel used for this research was obtained from the Department of Metallurgy and Material Science at Ahmadu Bello University, Samaru, Zaria. The mild steel composition by weight is: Fe (98.34%), C (0.08%), Si (0.26%), Na (0.64%), S (0.05%), Ni (0.09%), Cr (0.08%), Mn (0.02%) and Cu (0.27%). The mild steel sheet (0.8 mm thick) was mechanically cut into coupons of dimension 5.0 cm×1.0 cm for the weight loss measurement. For the electrochemical measurements, the coupons were cut to 1.0 cm by 1.0 cm coupons and then embedded in an epoxy resin giving an exposed surface area of 1 cm^2. The surface treatment of the samples was carried out by abrading the surface with different grades of emery paper from 600 to 1000 grit and then later degreased in ethanol, dried in acetone, and stored in a moisture free desiccator.

2.4. Gravimetric measurement

A pre-weighed mild steel coupon was completely immersed in 200 ml of the test solution in an open beaker. The beaker was covered with an aluminum foil and placed in a water bath to maintain a temperature of 303 K. After every 24 hrs for 144 hrs, the coupon was retrieved from the solution and the corrosion product was removed by washing each coupon with a solution of 50% NaOH, rinsed in acetone, air-dried and the weighed. This experiment was repeated at 303 K and 323 K for the blank and test solutions at different temperatures. The corrosion rate of mild steel (*CR*), fractional surface coverage (θ), and inhibition efficiency (*IE*%) were calculated respectively, using the Eq. (1).

$$CR = \frac{87.6W}{DAt} \tag{1}$$

Where W is weight loss in grams, D is density in gcm^{-3}, A is the area of specimen in cm^2 and t is the time of immersion in hours. The surface coverage (θ) and inhibition efficiency was calculated using Eqs. (2) and (3).

$$\theta = \frac{CR_{blank} - CR_{inhibitor}}{CR_{blank}} \tag{2}$$

$$IE = \frac{CR_{blank} - CR_{inhibitor}}{CR_{blank}} \times 100 \tag{3}$$

2.5. Electrochemical tests

Potentiodynamic polarization experiment was carried out using a conventional three-electrode electrochemical cell assembly. Freshly polished mild steel specimen with an exposed surface area of 1 cm^2 was used as a working electrode, saturated calomel electrode (SCE) as a reference electrode, and a platinum plate was used as the counter electrode. The measurements were performed using Gamry Electrochemical Analyzer. Potentiodynamic polarization curves were recorded by changing the electrode potential E_{corr} automatically with a scan rate of 0.5 mVs^{-1} from –0.25 to + 0.6 V_{SCE}. Before each run, the working and auxiliary electrode were immersed in the test solution for 30 minutes to attain a steady-state. The corrosion rate of the metal was calculated from the corrosion current density obtained from the extrapolation of the linear Tafel segment of anodic and cathodic curves. The inhibition efficiency

IE% was evaluated from the measured I_{corr} values as shown in Eq. (4) [23,24].

$$IE = \frac{I^0_{corr} - I_{corr}}{I^0_{corr}} \times 100 \quad (4)$$

Where, I^0_{corr} – corrosion rate in the absence of the inhibitor and I_{corr} – corrosion rate in the presence of the inhibitor.

3. Results and discussion

3.1. Effect of concentration and temperature

Mild steel corrosion behavior was investigated in the absence and presence of *Clindamycin* in 2.0 M H_2SO_4, using the gravimetric method at 303 K and 323 K. The variation of weight loss with immersion time for the corrosion of mild steel in the presence and absence of different concentrations of clindamycin at 303 K is shown in Fig. 2. In Fig. 2 it is observed that the weight loss of mild steel in the presence of clindamycin is lower than that of the blank solution. Meanwhile the weight loss decreased with the increase in the concentration of the clindamycin from 100 to 500 mg/L but increased with an increasing period of exposure. This indicates that the presence of the clindamycin retards corrosion of the mild steel in 2.0 M H_2SO_4 [25,26].

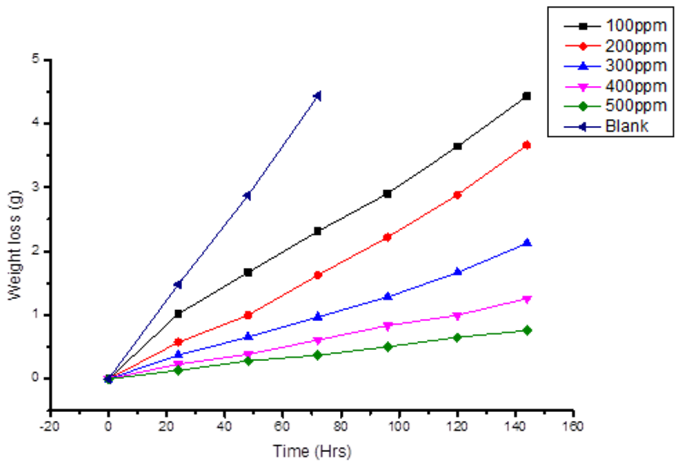

Fig. 2. Variation of weight loss with immersion time for corrosion of mild steel in 2.0 M H_2SO_4 containing different concentrations of clindamycin at 303 K

The corrosion rate was calculated for all the systems under investigation and the values obtained are were used to calculate the fractional surface coverage and inhibition efficiency as shown in Fig. 3 and TABLE 1. The corrosion rate of mild steel is noted to decreases with an increase in the concentration of clindamycin from 0.0604 mm/yr (100 ppm) to 0.053 mm/yr (500 ppm) at 303 K and from 0.4119 mm/yr (100 ppm) to 0.1236 mm/yr (500 ppm) at 323 K. This suggests that the clindamycin reduces the corrosion rate of the mild steel in 2.0 M H_2SO_4 both temperatures. Also, it is observed that the corrosion rates across the concentrations (100-500 mg/L of clindamycin) increased with an increase in temperature. This observation is in line with the expected increase in reaction rates as a result of increased thermal agitation [26,27]. The variations of the inhibition efficiency for different concentrations of the clindamycin at 303 K and 323 K are presented in Fig. 4. The inhibition efficiency and fractional surface coverage is noted to increase with the concentration at both temperatures, however, the inhibition efficiency at 303 K (with a maximum value of 91% at 500 ppm) was observed to be significantly higher than that at 323 K (with a maximum value of 70% at 500 ppm). The decrease in efficiency with increase in temperature suggests that the interaction between the mils steel surface and clindamycin may not be very strong; hence, an indication of a physical adsorption mechanism. In line with reports in the literatures, for a physical adsorption mechanism, the inhibition efficiency decreases with an increase in temperature while for a chemical adsorption mechanism, inhibition efficiency increases with the rise of the temperature [23-27].

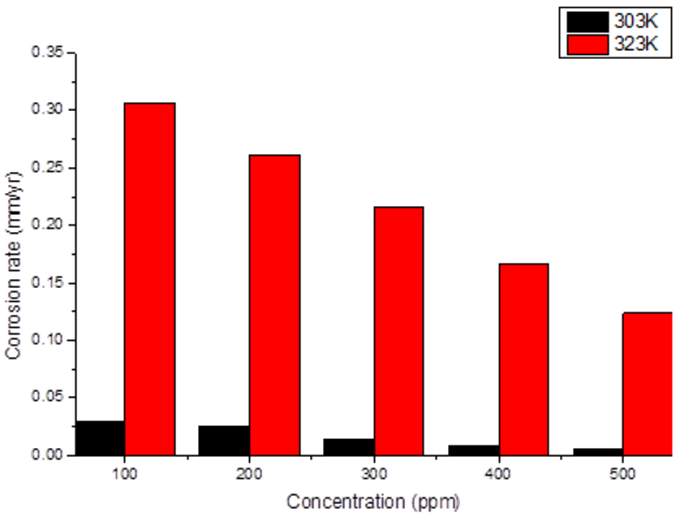

Fig. 3. Variation of corrosion rate with concentration of clindamycin drug 303 K and 323 K

TABLE 1

The corrosion rate of mild steel, inhibition efficiency and fractional surface coverage of the inhibitor 2.0 M H_2SO_4 at 303 K and 313K

Temperature (K)	Concentration (ppm)	Corrosion rate (mm/yr)	Inhibition efficiency (%)	Fractional surface coverage
303	Blank	0.0604	—	—
	100	0.0300	50	0.50
	200	0.0256	58	0.58
	300	0.0137	77	0.77
	400	0.0089	85	0.85
	500	0.0053	91	0.91
313	Blank	0.4119	—	—
	100	0.3069	25	0.25
	200	0.2614	37	0.37
	300	0.2164	47	0.47
	400	0.1668	60	0.60
	500	0.1236	70	0.70

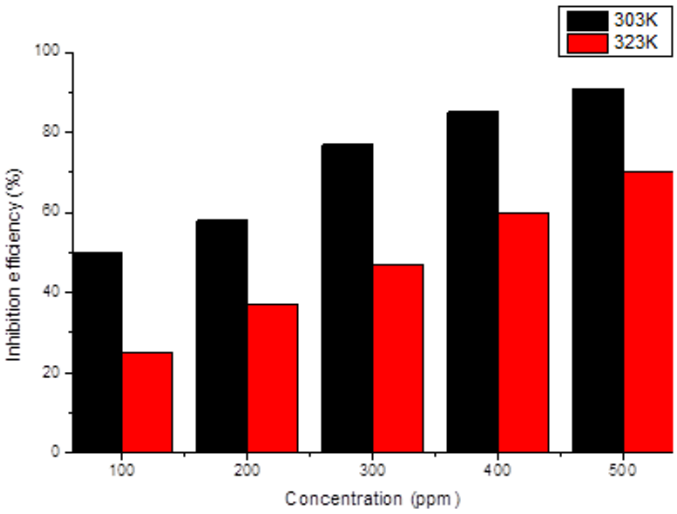

Fig. 4. Variation of inhibition efficiency of clindamycin drug with concentrations at 303 K and 323 K

3.2. Thermodynamic deductions

The activation energy for the corrosion of mild steel in 2.0 M H_2SO_4 in the absence and presence of clindamycin was calculated using the Arrhenius equation (Eq. (5)).

$$\log\left(\frac{CR_2}{CR_1}\right) = \frac{E_a}{2.303R}\left(\frac{1}{T_1} - \frac{1}{T_2}\right) \tag{5}$$

In Eq. (5), CR_1 and CR_2 are corrosion rates of mild steel at absolute temperatures T_1 (303 K) and T_2 (323 K), respectively, E_a is the activation energy for the reaction and R is the molar gas constant 8.314 J K^{-1} mol^{-1}). The heat of adsorption (Q_{ads}) for the clindamycin on mild steel was calculated using Eq. (6).

$$Q_{ads} = 2.303RT\left\{\log\left(\frac{\theta_2}{1-\theta_2}\right) - \log\left(\frac{\theta_1}{1-\theta_2}\right)\frac{T_1T_2}{T_2 - T_1}\right\} \tag{6}$$

Where θ_1 and θ_2 are the degrees of surface coverage of the inhibitor at temperature T_1 (303 K) and T_2 (323 K), respectively and R is the molar gas constant. The calculated values of activation energy, E_a and heat of adsorption Q_{ads} for the corrosion of mild steel surface in 2.0 M H_2SO_4 is shown in TABLE 2.

TABLE 2

Thermodynamic Parameters for the corrosion of mild steel surface in 2.0M H_2SO_4

Concentration (ppm)	E_a (kJ/mol)	Q_{ads} (kJ/mol)
Blank	15.56	—
100	18.70	–36.98
200	20.63	–39.56
300	23.64	–42.56
400	26.79	–47.12
500	29.80	–110.79

The calculated values of activation energy are shown in TABLE 2. Activation energy indicates the nature of the electrode kinetics, generally, the larger the activation energy the smaller the corrosion rate. From TABLE 2, the values of activation energy were observed to be higher than that of the blank solution (15.56 kJ/mol) meanwhile the E_a values increased with an increase in clindamycin concentration from 100 mg/L to 500 mg/L from a value of 18.70 to 29.80 kJ/mol. This observation indicates that additional amount of energy is needed by the system for corrosion to occur in the presence of clindamycin. Furthermore, for processes that occur by a physical adsorption mechanism, the activation energy is <80 kJ/mol while for chemical adsorption the activation energy is >80 kJ/mol [28,29]. In the present case, the activation energies are seen to be < the threshold values of 80 kJ/mol. This indicates that the adsorption of clindamycin on the mild steel surface is by physical adsorption. The values of Q_{ads} are also shown in TABLE 3, the heat of adsorption is a measure of the strength of interact between the adsorbate (clindamycin) and the solid adsorbent (mild steel). It is seen that the magnitude of Q_{ads} increased with increase in clindamycin concentration from –36.98 to –110.79 kJ/mol. This indicates that the strength of the interaction of clindamycin with the mild surface becomes stronger with the increase in clindamycin concentration.

TABLE 3

Langmuir adsorption parameters and free energy of clindamycin compound mild steel surface

T/K	Slope	R^2	log K_{ads}	K_{ads}	ΔG_{ads}/kJ/mol
303	0.8100	0.9864	–0.9450	0.9688	–10.03
323	0.7393	0.9768	–1.1169	0.8764	–9.52

3.3. Adsorption isotherm

The effectiveness of inhibitors may be explained by assuming that the inhibition process is as a result of adsorption. The extent of adsorption of different inhibitors at a fixed concentration would depend upon the surface area of the inhibitor molecules, the number of active centers such as N, S, and O atoms, and the intensities of lone pairs of electrons on these sites along with the intensities of pie-electrons on aromatic rings. The experimental data were tested with the Langmuir adsorption isotherm (Eq. (7)) and the plot is shown in Fig. 5. The plots of log(C/θ) versus log$C$ (Langmuir plot) yielded straight lines with correlation coefficient (R^2) and slope values given in TABLE 3 at different temperatures. The correlation coefficient and slope values in TABLE 3 are near unity indicating that the adsorption of clindamycin on steel obeys Langmuir adsorption isotherm [29-31]:

$$\log\left(\frac{C}{\theta}\right) = \log C + \log\left(\frac{1}{K_{ads}}\right) \tag{7}$$

Where K_{ads} is the adsorpton equilibrium constant, C is concentration of clindamycin and θ is the fractional surface coverage.

The values of K_{ads} at 303 K and 323 K temperatures were determined from the intercept lines on the log (C/θ) axis and listed in TABLE 3, together with ΔG_{ads} values for the inhibitor adsorption on the surface of mild steel, calculated using Eq. (8).

$$\Delta G_{ads} = -2.303\ RT \log(55.5 K_{ads}) \quad (8)$$

Where R is the molar gas constant, T is absolute temperature, 55.5 is the molar concentration of water in mol/L and K_{ads} is the equilibrium constant of adsorption defined as [30].

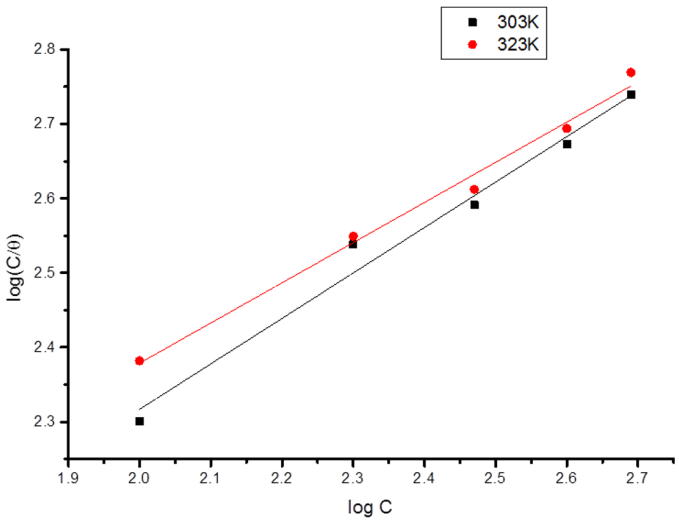

Fig. 5. Langmuir isotherm for the adsorption of clindamycin compound on mild steel surface

The magnitude of K_{ads} indicates the strength of the adsorbed layer [32,33]. From TABLE 3 it can be seen that the K_{ads} values are higher at 303 K (0.9688) than at 323 K (0.8764), this indicates that there is a stronger interaction between clindamycin and mild steel at 303 K compared to 323 K. The negative values of ΔG_{ads} indicate spontaneous adsorption of inhibitor molecules on the mild steel surface and strong interactions between inhibitor molecules and the metal surface. In general, values of ΔG_{ads} up to −20 kJ/mol are associated with physisorption, and those which are more negative than −40 kJ/mol are associated with chemisorption. The calculated ΔG_{ads} values for the clindamycin compound were found to be –10.03 kJ/mol and 9.52 kJ/mol at 303 and 323K, respectively, which fall within the region of physical adsorption, indicating that the adsorption process of inhibitors at mild steel surface involves physical adsorption.

3.4. Electrochemical test: Potentiodynamic polarization

Potentiodynamic polarization tests were carried out on the mild steel coupons in the presence and absence of clindamycin to deduce more kinetic parameters for the corrosion process. The potentiodynamic polarization curves of the corrosion of mild steel in 2 M H_2SO_4 solution in the absence and presence of clindamycin at 303 K is shown in Fig. 6. The addition of clyndamin is seen to have pronounced effect of the polarization behavior at lower and higher concentrations. The presence of clyndamin is observed to causes a change in E_{corr} values compared to that of the blank. The position of the Ecorr was below that of the blank solution at lower concentrations (100-300 ppm) while it was higher at 400 ppm. Fig. 6 also show that at lower concentrations (from 100-300 ppm) the cathodic current density decreased compared to that of the blank while there was no significant change in the anodic branch. At a higher concentration of 400 ppm, the anodic currents decrease compared to that of the blank. This shows that clindamycin behaves like a cathodic inhibitor at lower clindamycin concentrations and an anodic inhibitor at higher clindamycin concentrations. This indicates a possible change in the inhibition mechanism from a cathodic control process to an anodic controlled process on increasing the inhibitor concentration.

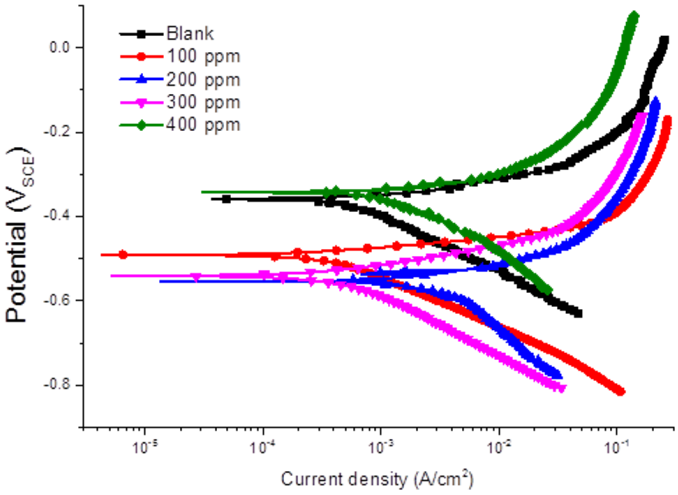

Fig. 6. Polarization curves of the corrosion of mild steel in 2 M H_2SO_4 solution in the absence and presence of clindamycin at 303 K

Furthermore, kinetic parameters were calculated such as E_{corr}, I_{corr}, cathodic (β_c) and anodic (β_a) Tafel slopes were calculated and listed in Table 4. The I_{corr} was calculated from the extrapolated linear portions of the curves from with β_c and β_a were deduced [34-37]. TABLE 4 shows that the E_{corr} values ranged from –347 to –565 mV. The calculated I_{corr} were noted to decrease from 1150 for the blank solution to 18 μA/cm^2 at 400 ppm. This result indicates that addition of clindamycin slows down the electrode kinetics and this effect increases as concentration of clindamycin increases. The β_c and β_a values were all lower than that of the blank (56.3 and 369 mV/dec^1 for β_c and β_a respectively). The results suggest the formation of a protective film on the mild steel surface which retards the electrode kinetics. This result is also consistent with the observation made from weight loss measurements and adsorption considerations. Hence, it is posited that clindamycin inhibits the corrosion of mild steel by physical adsorption of the steel surface, thus creating a barrier between the steel substrate and the corrosive medium.

4. Conclusions

Clindamycin has been evaluated for its corrosion inhibition performance on mild steel in 2.0 M H_2SO_4 solution, using gravimetric and potentiodynamic polarization techniques. The following conclusions have been drawn from the study.

1. Clindamycin inhibits the corrosion of mild steel in acidic media and the inhibition efficiency of the studied compound increases with increasing concentration
2. The corrosion mechanism is by physical adsorption of the clindamycin molecules on the steel surface and the adsorption of the clindamycin on a mild steel surface is an exothermic and spontaneous process that fits the Langmuir adsorption isotherm and involves a physical mechanism.
3. Potentiodynamic polarization results indicate that clindamycin behaves like a cathodic-type inhibitor at lower concentrations and an anodic-type inhibitor at higher concentrations.

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